(19) Japanese Patent Office (JP)

(12) Public Patent Disclosure Bulletin (A)

(11) Public Patent Disclosure Bulletin No.: 2000-186186 (P2000-186186A)

(43) Disclosure Bulletin Date: July 4, 2000

(51) Int. Cl.⁷	Identification Code	FI	Theme Code (Reference)
C08L 63/00		C08L 63/00	C 4F100
B32B 27/18		B32B 27/18	B 4J002
27/38		27/38	4J036
C08G 59/62		C08G 59/62	4M109
C08K 5/5397		C08K 5/5397	

Request for Examination: not yet submitted No. of Claims: 5 OL (Total Pages: 7)

---

(21) Patent Application No.: 10-364988

(22) Patent Application Date: December 22, 1998

(71) Applicant: 000002141
Sumitomo Bakelite Co., Ltd.
2-5-8 Higashi Shinagawa, Shinagawa-ku, Tokyo

(72) Inventor: Mikio Ito
2-5-8 Higashi Shinagawa, Shinagawa-ku, Tokyo (in-house)

(72) Inventor: Hiroshi Hirose
2-5-8 Higashi Shinagawa, Shinagawa-ku, Tokyo

To be continued on the last page

---

(54) Title of the Invention: Flame-retardant resin composition and the semiconductor sealing material and laminate which uses it

(57) Abstract

Problem: To propose a flame-retardant resin composition which has both a high degree of flame retardation without having to add any halogen compound and excellent reliability.

Means: It is a resin composition consisting of (A) an epoxy resin having at least 2 individual epoxy groups in one molecule without having to contain a halogen atom, (B) a hardening agent, and (C) an organic phosphorous compound having a structure in which there is a covalent bond of at least 1 phosphorous atom and a carbon atom in the molecule, and having at least 2 hydroxyl groups in the molecule. The phosphorous content in the resin composition is between 0.3 and 5% by weight.

Claims

Claim 1. A flame-retardant resin composition characterized such that it is a resin composition consisting of (A) an epoxy resin having at least 2 individual epoxy groups in one molecule and not having to contain a halogen atom, (B) a hardening agent, and (C) an organic phosphorous compound having a structure in which there is a covalent bond of at least 1 phosphorous atom and a carbon atom in the molecule, and having at least 2 hydroxyl groups in the molecule; and the phosphorous content in the resin composition is between 0.3 and 5% by weight.

Claim 2. The flame-retardant resin composition mentioned in *Claim 1* in which the hardening agent (B) is a compound or resin having at least 2 phenol type hydroxyl groups in 1 molecule.

Claim 3. The flame-retardant resin composition mentioned in *Claim 1* characterized such that the organic phosphorous compound (C) is the compound shown in the general formula (1).

[Chemical formula 1] ... (1)

where R_1 indicates a hydroxyl group or hydrogen atom.

Claim 4. A semiconductor sealing material which is basically composed of the flame-retardant resin composition mentioned in each of *Claims 1 ~ 3* and an additive.

Claim 5. A laminate which is basically composed of the flame-retardant resin composition mentioned in each of *Claims 1 ~ 3* and a backing.

Detailed Description of the Invention

[0001] Industrial Field of Application

The invention is concerned with a flame-retardant resin composition and the semiconductor sealing material and laminate which uses it which has excellent flame retardation without having to use a halogen-type flame retardant.

[0002] Prior Art

Thermohardened resins, represented by epoxy resins, etc., are widely used in electrical and electronic device parts because of their excellent characteristics and there are many examples which provide flame retardation thus insuring safety against fire. Generally in the prior art, compounds containing halogen of brominated epoxy, etc. have been used for making these resins fire-retardant. These halogen containing compounds have a high degree of flame retardation. Not only do aromatic bromine compounds separate corrosive bromine and hydrogen bromide in thermal decomposition, but, when breaking up in the presence of oxygen, highly poisonous polybromodibenzofurane and polybromodibenzooxine can be formed. Also, the treatment of waste and garbage containing bromine is very difficult.

[0003] For such reasons extensive research of phosphorous compounds as a flame retardant substituting for flame retardants containing bromine is a widely known fact. When using phosphoric acid esters in epoxy resins, however, the range in which they can be used in problems of bleeding and hydrolysis is limited. Also, with general phosphoric acid ester compounds having a functional group of a phenol type hydroxyl group, an separation of the phosphoric acid comes about by hydrolysis and the shortcoming arises of a marked deterioration in electrical properties and reliability.

[0004] Problems Which the Invention Is to Solve

This research is the result of assiduous investigation to solve the above mentioned problems and we are proposing a flame-retardant resin composition which has a high degree of flame retardation without having to add any halogen compound and the product characteristics of which do not deteriorate.

[0005] Means of Solving the Problems

Thus, the invention is a flame-retardant resin composition and the semiconductor sealing material and laminate which uses it characterized such that it is a resin composition consisting of (A) an epoxy resin having at least 2 individual epoxy groups in one molecule and not having to contain a halogen atom, (B) a hardening agent, and (C) an organic phosphorous compound having a structure in which there is a covalent bond of at least 1 phosphorous atom and a carbon atom in the molecule, and having at least 2 hydroxyl groups in the molecule; and the phosphorous content in the resin composition is between 0.3 and 5% by weight.

[0006] The Make-up of the Working Examples of the Invention

As mentioned above, when using phosphoric acid esters in epoxy resins, however, the range in which they can be used in problems of bleeding and hydrolysis is limited. Also, with general phosphoric acid ester compounds having a functional group of a phenol type hydroxyl group, an separation of the phosphoric acid comes about by hydrolysis and the shortcoming arises of a marked deterioration in electrical properties and reliability. In order to solve such problems in the present invention, the technical essence is that flame retardation and reliability coexist whereby, for a phosphorous compound to be generated having a stable P – C bond against hydrolysis resistance, by having at least 2 phenol type hydroxyl groups in the triphenylphosphine oxide structure, it should bond strongly with the resin matrix.

[0007] In the invention bisphenol A-type epoxy resins, bisphenol F-type epoxy resins, bisphenol S-type epoxy resins, phenol novolac type epoxy resins, cresol novolac type epoxy resins, naphthalene type epoxy resins, biphenyl type epoxy resins, N – glycidyl compounds from aromatic amine and a heterocyclic nitrogen base, for example, N, N – glycidyl aniline, trigycidayl isocyanurate, N, N, N', N' – tetraglycidyl bis (p – aminophenyl) – methane, etc. exemplify the epoxy resin (A) having at least 2 epoxy groups in 1 molecule and not containing halogen, but they [epoxy resins] are not particularly limited to these. Any kinds of these can also be used in combinations. However, as long as the invention advocates a resin composition which does not use a halogen type flame retardant, we exclude halogen containing epoxies such as and brominated bisphenol A epoxy and brominated novolac epoxy, but, in the manufacturing process of epoxy resins, chlorine, contained in ordinary epoxy resins originating from epichlorohydrin, is unavoidably intermingled. The quantity of that is of the order of several hundred ppm in the hydrolytic chlorine at a level generally known by the manufacturer.

[0008] For the hardening agent (B) used in the invention all those commonly known to manufacturers can be used but, particularly, straight chain diamines of C 2 ~ C20 such as ethylene diamine, trimethylene diamine, tetramethylene diamine, amines such as hexamethylene diamine, methaphenilene diamine, 4, 4' - diaminodiphenyl methane, 4, 4' - diaminodiphenyl propane, 4, 4' - diaminodiphenyl ether, 4, 4' –

diaminodiphenyl sulfone, 4, 4' – diaminodicyclohexane, bis (4 – aminophenyl) phenyl methane, 1,5 – diaminonaphthalene, metaxylilene diamine, paraxylilene diamine, 1, 1 – bis (4 – aminophenyl) cyclohexane, diaminodiamide, novolac type phenol resins such as phenol novolac resin, cresol novolac resin, tert – butylphenol novolac resin, nonylphenol novolac resin, etc., resol type phenol resin, polyoxystyrenes such as polyparaoxystyrene, phenol compounds in which the hydrogen atom is replaced with a hydroxyl group which bonds to a benzene ring, a naphthaline ring, another aromatic ring of naphthol type aralkyl resins such as phenol aralkyl resin (phenol resins and acid anhydrides such as are obtained by co-condensation of carbonyl compounds are good examples), but they [hardening agents] are not particularly limited to these.

[0009] As resin compositions for the semiconductor sealing material, the use of novolac type phenol resins such as phenol novolac resin, cresol novolac resin, tert – butylphenol novolac resin, nonylphenol novolac phenol resin, etc., resol type phenol resins, polyoxystyrenes such as polyparaoxytyrene, and naphthol type aralkyl resins such as phenol aralkyl resin are good because they are moisture proof and reliable.

[0010] Next, the organic phosphorous compound (C) used in the invention is shown in the general formula (1) and is synthesized by the method cited in *The Journal of Organic Chemistry*, 60, 3499 (1995).

[0011]

[Chemical Formula 1] ... (1)

where R_1 indicates a hydroxyl group or a hydrogen atom.

[0012] 4 – bromoanisol reacts with dichlorophenyl phosphine or trichlorophosphine and a phosphine oxide methoxy group substitute is obtained, then phosphine oxide hydroxyl group substitute is derived with tribromoboron.

PNF

[0013] The mixing proportion of the epoxy resin (A) and the hardening agent (B) can be established at will, considering those things in order for the (C) component to have a hydroxyl group. But, if any functional group has a large surplus, it is not good because of the decline in moisture proofing, moldability, and electrical properties of the hardened material. The proportion of the (C) component, the phosphorous content in the resin composition, i.e. weight proportion of the phosphorous atoms, should be in the range between 0.3 and 5% by weight. If this proportion is below 0.3% by weight there is no flame-retardant effect, and if it exceeds 5% by weight heat resistance and moisture proofing decline. Since this has an influence on moldability, it is not good.

[0014] The flame-retardant resin composition of the invention has a high degree of flame retardation without having to add any halogen compounds and is a thermohardened resin which will not cause deterioration of product characteristics. Beginning with sealings for semiconductor elements, it can be used aptly for sealings of electrical and electronics parts, coating, insulation, laminates, metal-clad laminates, etc.

[0015] As sealing materials for semiconductor elements and as filling, silica powder, alumina, talc, calcium carbonate, titanium white, clay, mica, etc. are combined as necessary; releasing agents such as natural waxes, synthetic waxes, straight chain aliphatic metallic salts, acid amide, ester, paraffin, colorants such as carbon black and red iron oxide, etc., and various hardening accelerators are used by manufacturers as commonly known additives and can be used as resin compositions for sealing. The flame-retardant resin

composition of the invention, additives and other things are chosen in a specific compositional ratio and, after mixing it so that it is sufficiently uniform by a mixer, etc., it is mixed with a heat roller or undergoes mixing treatment in a kneader, etc., it is cooled, made to harden, and pulverized into an appropriated size so that semiconductor sealing material is obtained. By transfer molding, injection molding, etc. it is used ideally for a sealing of a semiconductor element.

[0016] The varnish obtained by dissolving the flame-retardant resin composition in a solvent according to the invention is used in making coatings and laminates. For the making of a laminate, this varnish is applied to or impregnated upon a substrate of paper, glass woven fabric, glass unwoven fabric, or fabric not containing glass and by drying in a drying furnace in the range of 80 ~ 200 °C, a prepreg is prepared. Heating and pressing this, a metal-clad laminate for a laminate or printed circuit board is manufactured.

[0017] For the invention an ordinary solvent is used in the preparation of the varnish of the flame-retardant resin composition. It is necessary that the solvent used should show suitable solubility in part or the whole of the composition and a low-grade solvent can be used in a range where there will not be any bad consequences. If we were to give examples of solvents which can be used, they are ketone type solvents such as acetone, methyl ethyl ketone, methyl isobutyl ketone, cyclohexanone, etc., aromatic hydrocarbon solvents such as toluene, xylene, mesitylene, etc., glycol ether type solvents such as methyl cellosolve, ethyl cellosolve, butyl cellosolve, isobutyl cellosolve, diethylene glycol monomethyl ether, triethylene glycol monomethyl ether, propylene glycol monomethyl ether, dipropylene glycol monomethyl ether, propylene glycol monopropyl ether, ethylene glycol monoisopropyl ether, diethylene glycol monoisopropyl ether, diethylene glycol monoisopropyl ether, etc., ester type solvents such as methyl cellosolve acetate, ethyl cellosolve acetate, butyl cellosolve acetate, ethyl acetate, etc., dialkyl glycol ether type solvents such as ethylene glycol dimethyl ether, diethylene glycol diethyl ether, diethylene glycol dibutyl ether, etc. amide type solvents such as N, N – dimethyl acetoamide, N, N – dimethyl formamide, N – methyl – pyrrolidine, etc., and alcohol type solvents such as methanol, ethanol, etc. All kinds of these can also be used in combinations.

[0018] Below we will describe the invention in more detail giving working examples. First, we will refer to *Composition Examples 1 and 2* of the organic phosphorous compound which is an ingredient of the above-mentioned (C), then, we will refer to *Working Examples 1 ~ 5* in which the semiconductor sealing material and laminate are prepared combining these organic phosphorous compounds.

[0019] (*Composition Example 1*)

300 ml of dry THF and 3.78 g (1.5 mol) of magnesium flakes are put into a 3 L flask to which are attached an agitator, a dropping funnel, and a cooling tube; 7.5 ml of p – bromoanisol (60 mmol) is added. When the Grignard reaction has ended, 89.8 ml (0.72 mol) of p – bromoanisol is added for 1 hour. Then agitating it for 1 hour, 92.4 ml (0.39 mol) of dichlorophenyl phosphine is slowly dripped into it and, after dripping, it is agitated for 1 hour. Pouring this into a cold hydrochloric acid solution in which 10 g of ice have been put into 1 L of a 10% hydrochloric acid solution and ether extruding it, 75.1 g (75% yield) of bis (p – methoxyphenyl) phenyl phosphine is obtained.

[0020] 73.0 g (220 mmol) of bis (p – methoxyphenyl) phenyl phosphine is added to a solution of 37 g (230 mmol) of potassium permanganate in 2.5 L of water. This is agitated at room temperature for 16 hours, chloroform extruded, washed, and 66.9 g (90% yield) of bis (p – methoxyphenyl) phenyl phosphine oxide is obtained.

[0021] Putting 65.2 g (139 mmol) of bis (p – methoxyphenyl) phenyl phosphine oxide and 1 L of methylene chloride into a 2 L flask to which an agitator and cooling tube are attached, it is cooled in an acetone/dry ice solution to a temperature of – 78 °C. Putting 200 g (798 mmol) of 3 brominated boron into this, it is brought up to room temperature and agitated for 24 hours. Pouring it into 2 L of cold water, by heating the solvent is extracted and extruding in ethyl acetate, 40.9 g (95% yield) of bis (p – hydroxyphenyl) phenyl phosphine oxide (below, designated as the product (a)) was obtained. At a melting point of 232 °C by elemental analysis the phosphorous content was 9.9% and the hydroxide equivalent was 157 g/eq.

[0022] (*Composition Example 2*)

In the same manner as in *Composition Example 1* except for using 3 phosphorous chloride instead of dichlorophenyl phosphine, tris (4 – hydroxyphenyl) phosphine oxide (below, designated as the product (b)) was obtained. At a melting point of 273 °C by elemental analysis the phosphorous content was 9.3% and the hydroxide equivalent was 109 g/eq.

[0023] (*Working Example 1*)

We combined 80 parts by weight of fused silica, 10.7 parts by weight of YX – 4000H (biphenyl type epoxy resin, epoxy equivalence, 195 g/eq, manufactured by Shell Chemical Epoxy), 5.2 parts by weight of XL – 225 – 3L (Xyloc resin manufactured by Mitsui Chemical, hydroxyl group equivalence, 175 g/eq), 4.1 parts by weight of the product (a) obtained in *Composition Example 1*, 0.3 parts by weight of a separation agent (natural carnauba wax, 0.2 parts by weight of a pigment (carbon black), and 0.3 parts by weight of a coupling agent (A – 186 manufactured by Nippon Unica). Then, it is continually mixed using a heat roller and a molded material for use as a semiconductor sealing is obtained. We measured the spiral flow, hardenability, flame retardation, and reliability of this molded material. The spiral flow was measured under conditions of 175 °C, 70 kgf/cm^2 using a metal pattern based on EMMI 1 standards. Hardenability was evaluated by the molded product's buckle hardness for a melting time of 120 seconds at 175 °C. For the flame retardation test, after molding at 175 °C for 3 minutes and hardening it after 8 hours at 175 °C, a flame retardation test sample of 1.6 mm thickness is made and evaluated by the vertical method in accordance with UL 94 standards. Finally, for reliability, by the same molding conditions as in the flame retardation test sample, a monitor – IC (16pDIP) was molded upon which a simulated element was loaded, the reliability of this was evaluated with a failure number after leaving it for 1,000 hours at 125 °C and 100% humidity.

[0024] (*Working Examples 2 ~ 4*)

Except for mixing biphenyl type epoxy resin YX – 4000H, Xyloc resin XL –225 –L, and product (a) obtained in *Composition Example 1* or product (b) obtained in *Composition Example 2* according to Table 1, doing the same as in *Working Example 1*, the molded material was prepared and evaluated for spiral flow, hardenability, flame retardation, and reliability.

[0025] (*Comparison Examples 1 ~ 4*)

Except for combining each ingredient according to Table 2, the molded material is prepared in the same manner as in *Working Example 1* and its spiral flow, hardenability, flame retardation, and reliability is evaluated. The evaluation results of *Working Examples 1 ~ 4* and *Comparison Examples 1 ~ 4* are shown in Table 1 and Table 2, respectively.

[0026] Table 1

[Row 1, Column 1] Item

[Row 1, Columns 2 ~ 5] Working Ex. 1, Working Ex. 2, Working Ex. 3, Working Ex. 4

[Vertical heading for Rows 2 ~ 9] Combination Amount (Parts by Weight)

[Row 2] fused silica

[Row 5] product (a)

[Row 6] product (b)

[Row 7] natural carnauba wax

[Row 8] carbon

[Row 10] phosphorous component (% by weight in the resin composition)

[Vertical heading for Rows 11 ~ 14] Characteristics

[Row 11] spiral flow (cm)

[Row 12] hardenability (buckle hardness)

[Row 13] heat resistance test (UL 94)

[Row 14] reliability (failure product/sample)

[0027]

Table 2

[Row 1, Column 1] Item

[Row 1, Columns 2 ~ 5] Comparison Ex. 1, Comparison Ex. 2, Comparison Ex. 3, Comparison Ex. 4

[Vertical heading for Rows 2 ~ 10] Combination Amount (Parts by Weight)

[Row 2] fused silica

[Row 5] product (a)

[Row 6] resorcin diphenyl phosphate

[Row 7] triphenyl phosphine oxide

[Row 8] natural carnauba wax

[Row 9] carbon

[Row 11] phosphorous component (% by weight in the resin composition)

[Vertical heading for Rows 12 ~ 15] Characteristics

[Row 12] spiral flow (cm)

[Row 13] hardenability (buckle hardness)

[Row 14] heat resistance test (UL 94)

[Row 15] reliability (unsatisfactory product/sample)

[0028] In *Comparison Example 4* which uses a flame retardant of prior art, because the amount of flame retardant added is comparatively small, spiral flow is long, hardenability is low and reliability is also low in spite of a low flame retardation rating, V – 1. In *Comparison Example 3* which uses a flame retardant of prior art, by increasing the amount of flame retardant added, there was an improvement in flame retardation, V – 0, but reliability was lower. In *Comparison Example 1* which applied the organic phosphorous compound of the invention, because the amount of phosphorous compound combined, flame retardation was V – 1 and in *Comparison Example 2* where the amount of phosphorous compound combined was small and the hydroxyl group equivalence was excessive, reliability declined. In contrast to this, in *Working Examples 1 ~ 4* which applied the organic phosphorous compound of the invention, each one achieved a flame retardation rating of V –0 and, with the failure number of the monitor IC at 0, reliability was considered excellent.

[0029] (*Working Example 5*)

Adding the mixture solvent, methyl cellosolve/methyl ethyl ketone = 1/1, to 57.6 parts by weight of phenol novolac epoxy resin (Epicron) N-770 (a product of Dai Nippon, Inc., epoxy equivalence, 190 g/eq), 1.9 parts by weight of dicyan diamide, and 40.5 parts by weight of the product (a) obtained in *Composition Example 1*, a varnish is prepared to have a concentration of non-volatile matter of 50%. At this time, in 100 parts by weight total of the resin composition the phosphorous component becomes 4.0% by weight. Using this varnish, 80 parts by weight of the varnish solid are impregnated into 100 parts of glass fabric (thickness, 0.18 mm, Nitto Mills, Inc.), drying it for 4 minutes in a drying oven at 150 °C, a prepreg of 44.4% resin content is prepared. Layering 8 sheets of the prepreg obtained and on top of and underneath this a layer of 35 μm thick electrolytic copper foil, heat pressure molding is done for 120 minutes at 170 °C and 40 kgf/cm² and a laminate clad with copper on both surfaces of 1.6 mm thickness is obtained.

[0030] For the laminate obtained, flame retardation, solder heat resistance, peel strength, and glass transfer temperature are evaluated. Flame retardation is evaluated by a vertical method according to UL 94 standards. Solder heat resistance and peel strength are measured based on JIS C 6481, and for solder heat resistance, after carrying out a moisture absorption treatment by boiling for 2 hours, the existence of unusual outward appearance is investigated after it has floated in a solder tub at 260 °c for 180 seconds. Then, the glass transfer temperature is sought from the tanδ temperature by a visco-elasticity method.

[0031] *Comparison Examples 4 & 5*

Except for each ingredient being combined according to Table 3, a laminate was prepared in the same manner as in *Working Example 5* and flame resistance, solder heat resistance, peel strength, and glass transfer temperature were evaluated.

[0032]

Table 3

[Row 1, Column 1] Item

[Row 1, Columns 2 ~ 4] Working Ex. 5, Comparison Ex. 5, Comparison Ex. 6

[Vertical heading for Rows 2 ~ 5] Combination Amount (Parts by Weight)

[Row 2] Epicron N -770

[Row 3] dicyanamide

[Row 4] product (a)

[Row 5] triphenyl phosphine oxide

[Row 6] phosphorous component (% by weight in the resin composition)

[Vertical heading for Rows 7 ~ 10] Characteristics

[Row 7] flame resistance test (UL 94)

[Row 8] solder head resistance

[Row 8, col. 2 & 3] not unusual

[Row 8, col. 4] blistering occurred

[Row 9] peel strength (KM/cm)

[Row 10] Tg (DMA method · °C)

[0033] The evaluation results of *Working Example 5* and *Comparison Examples 5 & 6* were put together and are shown in Table 3. *Comparison Example* 6 which uses a flame retardant of prior art attained the heat retardation rating, V – 0, but, because the glass transfer temperature was low, in the solder heat resistance test blistering occurred and peel strength was also low. In contrast to this, for *Working Example 5* and *Comparison Example 4* solder heat resistance, peel strength, and glass transfer temperature all showed good results, but, although using the organic phosphorous compound of the invention, *Comparison Example 4* which has only a small amount added did not have satisfactory results in flame retardation.

[0034] Effects of the Invention

The resin composition using the organic phosphorous compound of the invention has a high degree of heat retardation without having to add a halogen compound combined with excellent reliability. It proposes a flame-retardant resin composition, and the semiconductor sealing material and laminate using it, which can be used suitably in applications where a non-halogen material is required in the future.

Continued from Page 1

(51) Int. Cl.⁷	Identification Code	FI	Theme Code (Reference)
H01L 23/29		H01L 23/30	R
23/31			

F Theme (Reference)

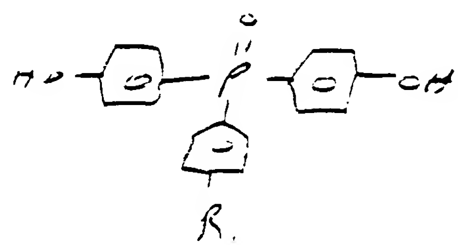
[refer to original (Page 7) for listing of numbers]

JP 2000 186186 A

FR Resin composition and the semiconductor sealing material and Laminate which uses it.

Claims

1. Resin consisting of
   a) epoxy resin ≥2 epoxy/molecule, no halogen
   b) hardening agent
   c) Org. P cmpd. w/ ≥ 1 P−C bond and ≥ 2 OH groups per molecule.
   And P content in resin is 0.3−0.5% wt
2. hardener is cmpd or resin ≥ 2 OH phenolic groups / molecule
3. Resin in claim 2 such that org P cmpd is



where R_1 = OH or H

4. Sealant matl.
5. Laminate composed of 1−3 and a backing.

C.Ex 1 Prep of CN2639 via PCl_3/BrAn Grig → $KMnO_4$ → BBr_3.

C.Ex 2. Prep of Tris via PCl_3 → $KMnO_4$ → BBr_3.

Working Ex 1. CN_2639 (4.1), other resins + fillers → Semiconductor sealants

" Ex 2−4 CN2639 or TrisOH + fillers → "

Comparison Ex 1−4 used CN2639 or resorcinol diphenylphosphate or TPPO

Working Ex 5. Phenol novolac DICY, CN2639 ⇒ prepreg → laminate V-0